Review of "FORest canopy atmosphere transfer (FORCAsT) 1.0: a 1-D model of biosphere-atmosphere chemical exchange"

Ashworth et al., GMD (2015)

Summary

This paper describes the development of a 1-D resolved canopy-chemistry model. FORCAsT is based on the well-established CACHE model but contains numerous updates, notably the incorporation of equilibrium gas-aerosol partitioning. Following a detailed description of the model parameterizations and updates to the chemical mechanism, comparisons to a few days of field data are presented for models runs with several different chemical mechanisms.

The manuscript is well written, though some re-organization might be considered as discussed below. The figures are appropriate but could use some improvement. The model description is thorough and includes a detailed supplement, making it appropriate for GMD. Publication is recommended after consideration of the following.

General Comments

Updates to the CACM mechanism: While the CACM mechanism is evidently not as good as RACM, it offers the advantage of coupling with an aerosol module; thus, a lot of time is spent fixing issues with CACM. While this is important, my concern is that Sect. 2.6.3 seems out of place as it is very long and involves analysis of field observations. It may be more appropriate to just give the important details in Sect. 2.6.2 and move most of this to the beginning of Sect. 4.

Also with regard to this subject, the titration of NOx by CACM0.0 is really striking. The lack of HO2 in this simulation is likely b/c of insufficient cycling via RO2 + NO, though this is not mentioned in the discussion. What is the primary source of NOx in the model? Advection? The accumulation of peroxy radicals as described in the last sentence of P. 5200 should shunt all of the NOx into NO2, but then where is the NO2 going? PAN? In short, it might be helpful to add a paragraph about the sources and sinks of NOx in the model.

Conclusions: a little time should be spent re-iterating what insights are gained from using the resolved canopy model versus using a 0-D model or a 1-D model without all the fancy canopy widgets. Hopefully the results presented in the last few sections can back up such statements.

Specific Comments

Sect. 2.4: Does the deposition scheme consider loss to surfaces other than leaves and soil (i.e. bark)?

Sect. 2.5: How is BL-FT exchange handled, and how is the height of the mixed layer determined?

P. 5197, L.15: "The similarity of the modelled concentrations suggest that differences in terpenoid oxidation pathways between the two chemistry schemes is of little importance compared to the magnitude of emissions and efficiency of vertical turbulent transport at this site." This sentence seems a little confused. Chemically, only the lifetime of primary emissions should affect their concentration, so they shouldn't care about subsequent chemistry except via feedbacks through radical cycling. Suggest restructuring to clarify what is meant here. There is a similar sentence at the top of P. 5210.

P. 5202, top: What is the assumed yield of isoprene nitrates? This information is not in the supplement (or at least I couldn't find it). If it is much larger than 12%, this would explain some of the remaining issues with the mechanism described later.

P. 5202, L. 13: The Muller (2014) paper is a theoretical study, not a lab study (though it does re-analyze some older lab experiments).

P. 5202, L. 24: ISOPO2 + HO2 does form carbonyls, but with a small yield (Liu et al., 2013).

P. 5203, 2nd paragraph: In most cases, 1st-generation isoprene hydroxyhydroperoxides should react with OH to form epoxides instead of photolyzing (the lifetime of ISOPOOH against OH reaction is a few hours). The omission of this pathway, which has been known since 2009 (Paulot et al., 2009), seems like a major shortcoming of CACM and RACM and could be problematic for low-NOx environments.

P. 5210, L. 3: In Fig. 4b, the model concentrations are at the low end of observations until noon of the second day.

P. 5211, last sentence: ozone has a larger reservoir and a long lifetime, so this isn't especially surprising. Likely, most of the ozone measured at UMBS was made upwind, so one would not really expect a 1-D canopy model to accurately capture ozone variability.

P. 2212, L. 9: Is the same data shown in Figs. 4i and 4j? If so, is it really fair to compare modeled HO2 to observed HO2*?

P. 5214, L. 10: Why do SOA concentrations maximize here?

Technical Comments

FIGURES: there are a lot of problems with the figures.

- For all figures, the font size should be increased. I had to magnify some to 300% to read them.
- Given that only 2 days are shown, it might be better to use hour-of-day, rather than day-of-year, as the x-axis coordinate.
- Fig. 1: what is the shading? What are the vertical lines?
- Fig. 3: Air temperature in C, not K

- Fig. 7: Might consider adding a dashed line showing height of mixed layer.
- Fig. 8: Might look better as cumulative-area plot

p. 5187: A few others that didn't make the list of canopy models are ACCESS (Saylor, 2012) and SOSAA (Zhou et al., 2014). The latter is particularly relevant as it is another canopy model with embedded aerosol mechanisms.

p. 5198, L. 4: It might be better to refer to the sum of MVK and MCR as "MVK+MCR."

Sect. 4.3: The discussion of the aerosol vertical profile might be better placed in terms of height relative to mixed layer depth rather than relative to canopy height. Or, just use absolute altitude. Using multiple height coordinates is confusing.

Supplement: There's a lot of info here (which is good!). A table of contents would be helpful.

References

Liu, Y. J., Herdlinger-Blatt, I., McKinney, K. A., and Martin, S. T.: Production of methyl vinyl ketone and methacrolein via the hydroperoxyl pathway of isoprene oxidation, Atmos. Chem. Phys., 13, 5715-5730, 2013.

Paulot, F., Crounse, J. D., Kjaergaard, H. G., Kurten, A., St Clair, J. M., Seinfeld, J. H., and Wennberg, P. O.: Unexpected Epoxide Formation in the Gas-Phase Photooxidation of Isoprene, Science, 325, 730-733, 2009.

Saylor, R. D.: The Atmospheric Chemistry and Canopy Exchange Simulation System (ACCESS): model description and application to a temperate deciduous forest canopy, Atmos. Chem. Phys. Disc., 12, 24765-24820, 2012.

Zhou, L., Nieminen, T., Mogensen, D., Smolander, S., Rusanen, A., Kulmala, M., and Boy, M.: SOSAA — a new model to simulate the concentrations of organic vapours, sulphuric acid and aerosols inside the ABL — Part 2: aerosol dynamics and one case study at a boreal forest site, Boreal Environment Research, 19, 237-256, 2014.